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Ferromagnetic resonance linewidth reduction in Fe/Au multilayers using ion beams

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In order to optimize their magnetic properties, Fe/Au multilayers were treated by pregrowth and postgrowth ion-beam bombardments. The ferromagnetic resonance linewidth was used as our main figure of merit. The pregrowth treatment of the MgO substrate using a 60 eV atomic oxygen beam resulted in a reduction of the inhomogeneous linewidth broadening in comparison with a sample grown on an untreated substrate. This homogeneity increase is linked to the removal of substrate carbon contamination by the chemically active oxygen. It correlates with the reduced interface roughness. The postgrowth sample irradiation using 30 keV He⁺ ions also reduces the inhomogeneous broadening in the linewidth. Fe and Au have a miscibility gap, but the demixing is kinetically quenched at room temperature. Ion collisions locally minimize the interface energy by providing the energy necessary for localized demixing, resulting in a smoothing effect. Combined, the pregrowth and the postgrowth irradiations lead to the lowest observed linewidth. © 2008 American Institute of Physics. [DOI: [10.1063/1.2839266](https://doi.org/10.1063/1.2839266)]

Engineering the magnetic properties of thin films and multilayers is imperative to reveal novel effects or optimize the functioning of devices. In particular, in the latter, most often ultrathin layers are employed. Due to the reduced dimensions, the interface properties, e.g., its roughness,¹ gain considerable importance. While optimizations of the growth procedure by, for example, using buffer or seed layers² are possible, ion irradiation constitutes an alternative method, which can be used in addition. Two different treatments of Fe/Au multilayers are assessed in this article. A pregrowth ion-beam treatment of the MgO substrate using atomic oxygen: cleaning the substrate in this manner promises improved growth on top. Postgrowth irradiation using He⁺ ions: while ion irradiation is often associated with mixing, this is not the case here. As Fe and Au are thermodynamically immiscible, we assume that the energy transferred by the ion collisions should make localized demixing at the interfaces possible. Finally, we analyze if both treatments can be combined to achieve an even more pronounced effect.

Two Fe/Au multilayer samples were grown by molecular beam epitaxy. Their nominal structure is MgO(100)/Fe(3)/Au(19)/[Fe(1)/Au(0.9)]₂₀. All thicknesses are expressed in nanometers. The 1 × 1 cm² sized MgO substrates were mounted on a rotating molybdenum plate to improve homogeneity. Prior to growth, they were annealed for 1 h at 500 °C removing water and hydrogen. The 3 nm Fe seed layer was deposited at the same temperature. It leads to an improvement of the growth conditions for

the subsequent 19 nm Au buffer layer,^{2,3} which was deposited at 200 °C. A surface roughness of the Au layer on the order of the interlayer spacing is expected.⁴ The actual Fe/Au multilayer was deposited at 70 °C to minimize interdiffusion of the alternating layers. The growth pressure was 6.9 × 10⁻⁹ Torr. The sample A-non was grown on a MgO substrate, which was additionally pregrowth ion beam treated. The procedure consists of an exposition for 2 min to a neutralized atomic oxygen beam with the nominal energy of 60 eV, providing an ion dose of 6 × 10¹⁶ ions/cm². While the substrate was irradiated in vacuum (10⁻⁴ mbar), it was kept in air until the growth in the molecular beam epitaxy system. It was shown by Auger spectroscopy that the treatment removes the carbon contamination, which is found on the surface of MgO substrates.³ The second sample, B-non, was deposited on a substrate only subject to the described annealing procedure.

The samples were characterized using the vector network analyzer ferromagnetic resonance (FMR): the thin films are placed on a coplanar waveguide, which creates a small oscillating field perpendicular to a constant magnetic field applied by an electromagnet. A small angle precession is excited, which is detected using the network analyzer. A detailed description of the measurement procedure is found elsewhere.⁵ Varying the frequency of the small excitation field, the measured data allow the calculation of the dynamic susceptibility spectra of the sample. This is illustrated in Fig. 1, plotting the imaginary part of the susceptibility of both samples, under an applied field of 45 mT.

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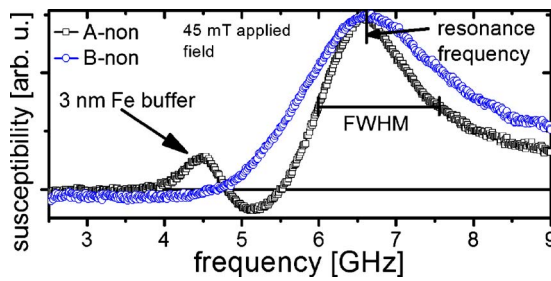


FIG. 1. (Color online) Imaginary part of the complex susceptibility of the A-non and B-non samples for the same applied field of 45 mT. The linewidth is broader for the B-non sample. The small second resonance is only visible for the A-non sample.

From each spectrum, we evaluate two quantities: the FMR frequency f_{res} and the full linewidth at half maximum Δf . The resonance frequencies almost coincide. They allow to determine the effective magnetization using the appropriate equation for the resonance condition.⁶ We find $\mu_0 M_{\text{eff}} = 0.97$ T for the A-non sample and 0.99 T for B-non, where we assumed a g factor of $g = 2.1$. These values are confirmed by conventional magnetometry. They are in agreement with expected values for the effective magnetization of 1 nm thick Fe layers, as the saturation magnetization is reduced by a surface anisotropy contribution.⁷

The resonance linewidths differ considerably. To permit an interpretation, Fig. 2 plots Δf as a function of the external applied field H_{appl} .

The linewidths clearly exceed the values observed on single Fe layers. This is attributed to the multilayer, where the measured linewidth is extracted from a superposition of resonances of all individual layers, which are slightly shifted due to different effective fields. Therefore, the extracted properties can only be related to the ensemble of layers. The linewidth and the intrinsic Gilbert damping parameter α are related as

$$\Delta f_{\text{int}} = \alpha \frac{\gamma \mu_0}{2\pi} [M_{\text{eff}} + 2(H_{\text{appl}} + H_{\text{aniso}})], \quad (1)$$

where γ is the gyromagnetic ratio and H_{aniso} the anisotropy field. The expected curve for Δf_{int} is included in Fig. 2 (dotted line). The measured curves deviate in shape from Δf_{int} .

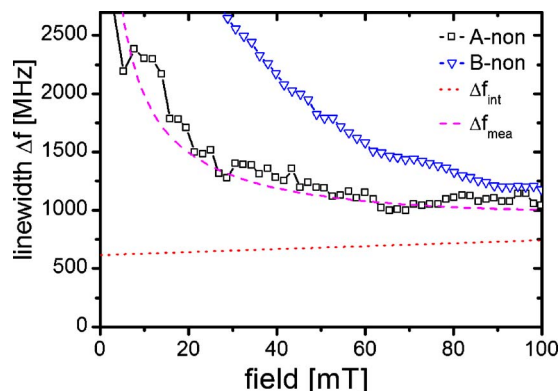


FIG. 2. (Color online) Resonance linewidth as a function of the applied magnetic field for the A-non and B-non samples. A fit including the extrinsic linewidth contribution is shown for A-non. The intrinsic linewidth using the fitted damping parameter is plotted.

This is related to the inhomogeneous broadening: this phenomenon is well known in field-swept FMR,⁸ where it is observed as a finite zero-frequency field linewidth ΔH_{inhomo} . In frequency-swept FMR, on the other hand, the extrinsic frequency linewidth Δf_{inhomo} is not constant with resonance frequency but decreases with its inverse. The analytical description⁹ yields $\Delta f_{\text{inhomo}} \propto \Delta H_{\text{inhomo}} / f_{\text{res}}$. It adds in quadrature to the intrinsic linewidth,¹⁰

$$\Delta f_{\text{mea}}^2 = \Delta f_{\text{int}}^2 + \Delta f_{\text{inhomo}}^2. \quad (2)$$

Equation (2) allows to separate the extrinsic contribution in Fig. 2: α was determined to be 0.022. It is difficult to quantitatively attribute Δf_{inhomo} to the spread in one magnetic property. However, it is reasonable to assume that a major contribution to the inhomogeneity is related to roughness, as the individual magnetic layers in the sample consist of only few atomic layers. A qualitative comparison of the extrinsic linewidth contributions confirms a higher homogeneity in the A-non sample grown on the cleaned substrate than in the B-non sample. Atomic force microscopy images on the surface of the uppermost Au layer support the connection between the lower interface roughness and the higher homogeneity: the root mean square roughness was determined to be 0.27 nm for the A-non sample and 0.44 nm for B-non. The effect of the substrate cleaning can thus still be observed after a stack of 42 layers.

Another strong indication for the better growth conditions on the cleaned substrate is the observation of a second magnetic resonance in the spectrum of the A-non sample (see Fig. 1). It is caused by the lowermost 3 nm thick Fe seed layer. From its resonance frequency, we obtain $\mu_0 M_{\text{eff}} = 1.5$ T. We could not observe this resonance for B-non: grown on the rougher substrate, it most likely has a magnetic dead layer, its signal too low for detection.

An alternative means of influencing the layer quality is the postgrowth light-ion irradiation.¹¹ The irradiation parameters were determined from stopping and range of ions in matter SRIM simulations:¹² 30 keV He⁺ ions lead to a nuclear stopping power of 1.28 eV/Å in Fe. This low value maintains the crystallographic structure, typically displacing single atoms only a few interatomic distances. Though Fe and Au are immiscible, demixing of the FeAu alloy at the multilayer interfaces is kinetically quenched at room temperature. The ion collisions provide the atom mobility to overcome this kinetic limit, yielding an effective interface smoothing effect. Due to the immiscibility, it is thermodynamically favorable to reduce the interfacial area and the corresponding interfacial energy.¹³ The penetration depth of the incident ions is estimated to be 113 nm, i.e., they remain in the substrate.

The two initial samples were diced into several pieces. They were irradiated with 30 keV He⁺ ions at a low current of 0.25 $\mu\text{A}/\text{cm}^2$, avoiding sample heating. A-low and B-low are pieces of the respective nonsamples irradiated with a fluence of 1×10^{15} ion/cm². A-high and B-high represent pieces of the initial samples irradiated with a fluence of 3×10^{15} ion/cm².

Figure 3 shows the linewidths of the samples B-non, B-low, and B-high at 45 mT applied field, centered around 0

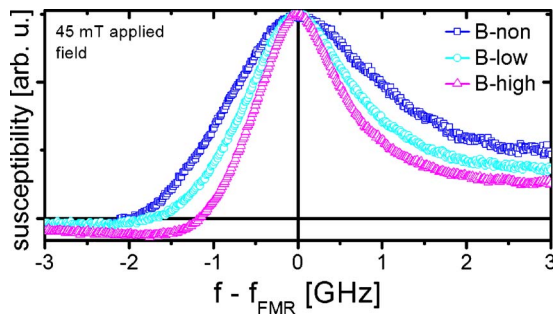


FIG. 3. (Color online) The imaginary part of the complex susceptibility of the three B samples for the same applied field of 45 mT centered around 0 (shifted by their respective resonance frequency). The linewidth is smaller for a higher irradiation dose.

for better comparison. The irradiation led to a slight increase in effective magnetization, yielding a higher resonance frequency for the same field. This increase is not understood.

The linewidth decreases when irradiating with a higher fluence. Figure 4(b) plots the measured linewidths for the B samples as a function of applied field. The lower linewidth for the irradiated samples is attributed to a lower inhomogeneous broadening. In analogy to the preceding argument, the lower extrinsic linewidth can be related to a lower interface roughness.

Indeed, the lowest inhomogeneous broadening is observed for the A-high sample in Fig. 4(a). This sample was deposited on the pregrowth ion-beam cleaned MgO substrate

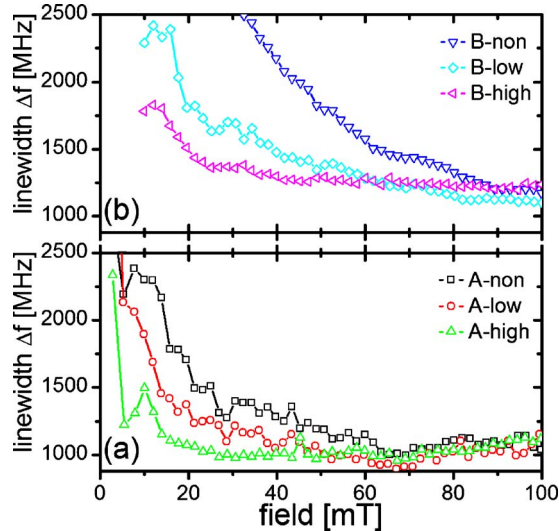


FIG. 4. (Color online) Resonance linewidth as a function of the applied magnetic field for the three A samples (a) and for the three B samples (b). At lower fields, the linewidths are clearly larger for the B samples compared to the A samples.

and was postgrowth irradiated with the higher fluence. We conclude that the smoothing effect of both methods add up. The efficiency of the postgrowth treatment still has room for improvement by further augmenting the fluence, as defect creation is omitted due to the low ion energy.

In summary, we have shown that it is possible to improve the interface roughness of Fe/Au multilayer samples by ion-beam irradiation. A pregrowth substrate treatment using a dissociated 60 eV atomic oxygen ion beam removes the carbon contamination on MgO substrates due to its high chemical activity. This allows us to decrease the interface roughness of all subsequently deposited layers. The procedure is even still effective after exposing the substrate to air for a longer period. The postgrowth irradiation using 30 keV He⁺ ions also improved the interface smoothness. The low energy avoids defect creation. The ion collisions provide the atom mobility necessary to achieve localized demixing of the FeAu alloy at the interfaces. Increasing the irradiation fluence decreases the inhomogeneous linewidth broadening. The pregrowth and postgrowth treatments combined lead to the lowest extrinsic broadening, i.e., the highest homogeneity.

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